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Soft triboelectric generators by use of cost-effective elastomers and simple casting process

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ABSTRACT

We are reporting a new approach of developing of soft triboelectric devices with improved conformability and scalability. Here, elastomers, namely, polydimethylsiloxane (PDMS), polyurethane (PU), were used as triboelectric layers, and their carbon based conductive elastomeric composites were implemented as electrodes since they offer greater softness and flexibility. Elastomers can also be coupled with the textile. Simple film casting technique, that provides fast and readily scalable route, was used to prepare low-cost multilayer structures. The technology was tested by preparing vertical-contact separation based triboelectric shoe-insole and stretchable triboelectric band. These examples were tested at low frequencies and at different load resistors for different gestures. The shoe-insole generated optimum rms power (P_{rms}) of 0.28 m W for load resistance (R_L) of 150 M Ω for walking and 1.79 m W for the R_L of 110 M Ω for running, respectively. Stretchable band, which can be applied on different body joints, was tested for the arm stretching motion that delivers the optimum P_{rms} of 9 μ W for the R_L of 400 M Ω.

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1. Introduction

The fast development of the internet of things (IoT) and different types of consumer low power electronics and sensors, prompted the requirement of portable power source. The conventional power supply, such as, batteries, has limited lifespan that requires frequent monitoring and replacements, and hazardous for the environment due to use of various chemicals. Therefore, with the ultimate aim of developing fully autonomous electronic systems, over the past years, growing demand for environment friendly, maintenance-free, sustainable, light weight and cost-effective renewable power sources are observed.

Like other renewable sources, different types of mechanical and kinetic energies can be a potential prospect to power energy-efficient electronic devices [1,2]. Generally, electromagnetic, electrostatic and piezoelectric techniques are used to convert mechanical energy to electrical signal [2–4]. In recent time, triboelectric effect, which is mainly known for its negative effects in electronic industries for years [5], was reported to be utilized for harnessing and sensing of mechanical energies [6].

Triboelectric nanogenerator (TENG) basically uses contact electrification and electrostatic induction methods. The physical contact or friction between two materials, having opposite charge affinity [7], is required for this mechanism to be functional. Since its discovery in 2012 [6], TENGs have shown exciting prospects for energy harvesting at large and small scale from surrounding kinetics energies, and for pressure/motion sensing applications [8–17]. In addition, triboelectric effect allows energy scavenging from and/or sensing of low frequency motions, such as, human body movements [10,18]. Wide variety of materials can be used to fabricated such devices [7]. Although, polytetrafluoroethylene (PTFE) is generally preferred as triboelectric materials due to its highest affinity toward negative charges, but this material is expensive, it is not standard in the field of microfabrication, adhesion of other materials on PTFE foil, such as the charge collector electrodes, is problematic. Furthermore, for wearable systems, materials need to be soft and flexible, which make them adaptive to various complex surface topologies and enable them to withstand high mechanical deformations.

To date, various triboelectric nanogenerators have been prepared and reported, with the ultimate goal of developing fully autonomous wearable electronic systems [10,19–29]. As an example, fabrication of triboelectric shoe insoles [19,20] have been reported to harvest energy for walking and with the capability of charging consumer electronics. The stretchable rubber based tri-

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boelectric device with the signal electrode has been reported [21], where the in-plane charge distribution due to the stretching of the rubber led to the charge flow between electrode and ground. Such system can be used to detect moving direction. In addition, a conductive fabric based stretchable single electrode TENG was demonstrated [22]. The friction between the dielectric layer and carbon nanotubes based custom made conductive fabric generated the electrical signal. Furthermore, stretchable fibre based triboelectric nanogenerators have been described [23], where copper wire convolved the silicone fibre structures with stretchable electrode. Kim et al. [24] have reported vertical contact separation mode based highly stretchable wearable TENG, where a metallic wire was placed inside the PDMS tube. However, there are still lack of simple technological approach that combines cost-effective processing technique and use of economical materials to fabricate low-cost soft triboelectric devices, which is compatible with large area applications and can be readily industrialized.

This work presents, the fabrication of soft triboelectric devices with improved conformability, scalable, and cost-effective process. Soft elastomeric materials were used, during this work, because of their higher softness and flexibility. In addition, the elastomers used are biocompatible, which make them more preferable for developing wearable devices, and ultimately implantable devices. Here, soft dielectric elastomers, namely, PDMS and PU, were employed as triboelectric layer, and their carbon based conductive composites were utilized as electrodes. Moreover, we demonstrated that the elastomeric technology can be incorporated with textile layers. The simple film casting technique was adapted to pattern multilayer structures of various materials, which is fast and applicable for large area applications. Finally, to manifest the potential of this technology, evaluation of two different soft triboelectric configurations, such as, vertical-contact separation and stretchable triboelectric devices, in the forms of shoe insole and stretchable band are presented as examples. Both the generators have simple designs and were characterized at low frequencies for different gestures.

2. Soft elastomers and casting approach

Materials explored herein, during this development process, were elastomers, as they are low-cost, and provide, thanks to their stretchability, conformability and softness, and they can be biocompatible as well. Elastomeric materials can be patterned for variable area and geometries, and can easily be adapted and optimized for large area manufacturing and nowadays for 3D-printing. Furthermore, elastomeric materials can be combined with various electrically conductive particles to develop elastomeric electrode materials, which can be casted directly on various material layers and provide better adhesion between dielectric and conductive layers without compromising much of their mechanical properties. Finally, these functional materials can be solution processed and patterned using the simple film casting technique. The latter allows to deposit materials having wide range of viscosities with different thickness. Therefore, the use of elastomeric materials along with the casting approach provides cost-effective and simple technological platform, which can be applied for the fabrication of conformable devices with different geometries and areas.

In this work, soft dielectric elastomers widely available, namely, polydimethylsiloxane (PDMS) and polyurethane (PU), were exploited as functional materials. These materials have opposite charge affinities that promoted contact electrification and thus can be used to prepare triboelectric devices. Moreover, by adding to them cost effective carbon black particles elastomeric carbon-based PDMS (CPDMS) and carbon-based PU composites (CPU) conductive composites were prepared for the electrodes of the generators. Furthermore, these elastomers can also be directly casted

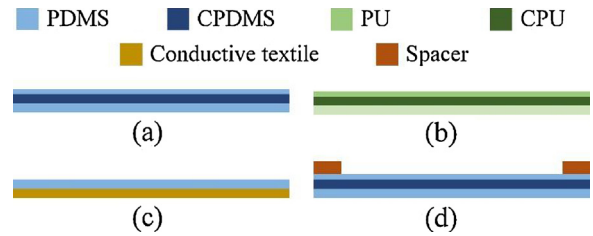

Fig. 1. Schematic diagram of the layer structures (a) PDMS-CPDMS-PMDMS, (b) PU-CPU-PU, (c) PDMS-textile and (d) PDMS-CPDMS-PDMS layer with spacer.

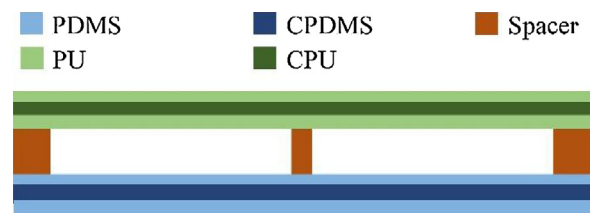

Fig. 2. Schematic diagram of the design of vertical mode triboelectric generator implemented as shoe insole.

on or integrated with the electronic textile (e-textile), which enable easy incorporation of soft triboelectric generators with clothing.

The technological realization processes began with the development of elastomeric PDMS, PU and their composites materials preparation and optimization of the casting processes. The triboelectric structures were made of three casted layers. The initial PDMS layer acts as protective/ support layer, followed by the casting CPDMS layer as electrode and final layer of PDMS as active triboelectric layer. As result, we obtained PDMS-CPDMS-PDMS layer structure (Fig. 1(a)). Similarly, triboelectric PU structure was developed by casting PU, CPU and PU layers to obtained PU-CPU-PU structure, as shown in Fig. 1(b).

The integration of the PDMS elastomer with an electrically conductive stretchable textile materials was also examined. This can be performed either by casting the PDMS layer directly on the textile or by laminating approach. It has been observed that the direct casting of PDMS could reduce the stretchability of the layer due to materials absorption by the fabric. Therefore, lamination process was optimized during this work. In this regard, ultra-thin layer of PDMS elastomer, acting as adhesive, was casted on the previously prepared PDMS layer, and then electrically conductive textile was laminate with the PDMS layer by applying pressure. The resulting structure provides PDMS-textile layer (Fig. 1(c)).

Moreover, in case of requirement, elastomers can either be casted or molded directly on the active PDMS layer to create spacer, as presented in Fig. 1(d). In this case, stencil is used to cast the spacer shape on active PDMS layer. The potential of the developed technological platform was tested by preparing triboelectric devices applying vertical-contact separation and vertical-contact separation/sliding mechanisms. As examples, we realized triboelectric generator based shoe insole and stretchable band. Their designs, operation principles and the results obtained on these devices are discussed in the later sections.

3. Experimental procedures

3.1. Designs & materials

The vertical contact mode based soft triboelectric device was tested by preparing the form of shoe insole. Fig. 2 presents the cross-sectional view of the design of the triboelectric insole. The fully elastomeric vertical contact mode generator was composed of two active layers separated by a spacer and assembled facing

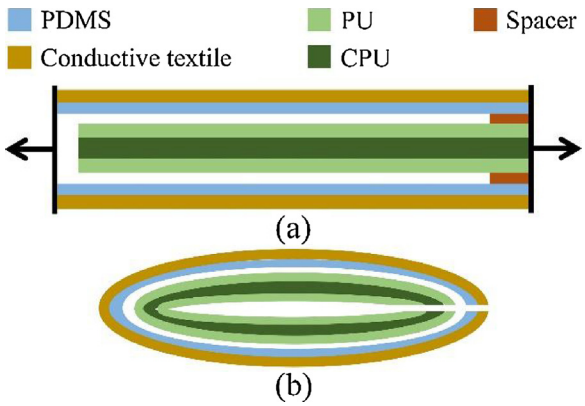

Fig. 3. Schematic diagram of the design of the stretchable triboelectric band along the (a) length and (b) width directions.

one another. PDMS and PU were used as triboelectric layers and their respective carbon-based conductive elastomeric composites were used as electrode materials. The spacer was made of thick soft PDMS material, which acted as spring during the application and relaxation of the mechanical force to generate the contact and separation of the triboelectric layers. The repetition of the walking or running motions create contact-separation cycles of the two triboelectric layers and thus provides harmonic electrical output responses.

A stretchable triboelectric generator that combines the vertical contact-separation and sliding modes mechanism was realized using the soft materials integrated to a commercially available stretchable electrically conductive textile. The proposed design for the stretchable band was formed by putting together two triboelectric layers (PDMS and PU) facing each other separated by thin spacer between them. The inner layer was fixed with the outer layer only at one end and the other end of the inner layer is free to move between the outer layer, as shown in Fig. 3. The outer layer is stretchable and made of conductive textile functionalized with a PDMS triboelectric layer, whereas the inner layer is non-stretchable but flexible and was composed of thin PU layer with electrically conductive carbon-based elastomer electrode. The conductive textile provides better conductive electrode, mechanical properties and easy integration with the clothing without damages. Furthermore, the use of conductive textile as electrode for the outer stretchable layer helps to hold the surround elliptical shape, as shows in Fig. 3, without touching the inner layer due to the elastic properties of the textile itself. The spacer is made by attaching thin stretchable PDMS layer on active PDMS triboelectric layer using elastomeric adhesive. While movement, stretchable band mounted on body joint, both lateral-sliding and vertical contact separation principles acted simultaneously. The stretching lead to variation of the overlapping surface area due to sliding of the inner layer inside Stretchable PDMS layer. In addition, stretching caused changes in the charges distribution on the stretchable PDMS surface.

During this work, materials were used as received. Two parts biocompatible Sylgard 186 silicone (PDMS) elastomer kit, and biocompatible MM4520 polyurethane (PU) were purchased from DowCorning and SMP Technology Inc., respectively. The ketjenblack EC-300J carbon-black pellets that being used to prepare conductive carbon-based elastomers composites, were shopped from Akzo-Nobel, Japan. MedTex180 silver-plated electrically conductive nylon fabric (MedTex180) was purchased from Shieldex productions & Vertriebs GmbH. Sigma-Aldrich supplied the solvents and polyethylene terephthalate (PET) foils were ordered from commercial source.

Table 1
List of physical parameters of the triboelectric generator based shoe insole and stretchable band.

Physical parameters	Shoe insole	Stretchable band
Total area [cm^2]	133	72
Thickness of PDMS layer [μm]	60	60
Thickness of PU layer [μm]	50	50
Spacer thickness [mm]	2.0	0.2

3.2. Process flow

PDMS was prepared by mixing two parts and PU were dissolved into solvents prior to use. For the electrodes materials, 10 wt.% of electrically conductive carbon black particles were dispersed into elastomer matrices to prepared CPDMS and CPU composites. Film casting was performed using film applicator. Prior to the casting of functional layer sacrificial layer was casted and dried on PET substrate at room temperature. During the process, drying and curing steps for PDMS, PU and their composites were performed at 80 °C for 2 h. The spacer for the vertical contact separation mechanism based shoe insole was developed by casting PDMS using stencil directly on the PDMS-CPDMS-PDMS structure and cured.

Once prepared, layers were assembled in accordance with the design to devise vertical contact separation based triboelectric shoe insole and stretchable triboelectric band. During assembly process, PDMS layer structures were released from substrate by dissolving sacrificial layer. The shoe insole was prepared using PDMS-CPDMS-PDMS and PU-CPU-PU structures, where active PDMS and PU layers were facing one another separated by casted spacer. Fig. 4(a) exhibits the photograph of the top view of the triboelectric shoe insole.

Similarly, PDMS-textile and PU-CPU-PU layers, respectively, as outer and inner layer, were used to develop stretchable triboelectric band. The assembly process of the stretch band was performed by mounting and fixing one edge of the inner layer with the one end of the stretchable outer PDMS-textile layer and finally folding the layers together, as shown in design. In this case, thin stretchable spacer layer was used to separate the two active layers to avoid any electrical short circuit. The photograph of the stretchable triboelectric band is presented in Fig. 4(b). The physical dimensions of the developed triboelectric generator based shoe insole and stretchable band are tabulated in Table 1.

3.3. Characterizations

During this work, triboelectric devices, both shoe insole and stretchable band, were characterized on body of a person having a weight of 76 kg, for different movements. The electrical responses across range of load resistances were obtained using the oscilloscope, and the corresponding current outputs and powers were calculated. Fig. 5(a) illustrates the circuit diagram of the setup. The open circuit voltage (V_{OC}) was measured across resistance value of 11 G Ω; whereas maximum current output (I_{Max}) was measured over 1 M Ω resistance value, since the responses for the load resistances below 1 M Ω were too noisy for proper detection. In addition, the amount of charges transferred and energy stored of the triboelectric devices under mechanical motions on body were determined by storing charges in a capacitor (C_L) of 2.2 μ F, as showed in Fig. 5(b), through full wave bridge rectifier for ac-to-dc signal conversion.

Shoe insole was characterized while walking and running motion at 0.65 ± 0.10 Hz and 1.60 ± 0.10 Hz, respectively. For the stretchable band, the initial characterizations were performed under stretching for the strain of 10% and tapping motions, where the operation frequency of 0.65 ± 0.15 Hz was maintained for both

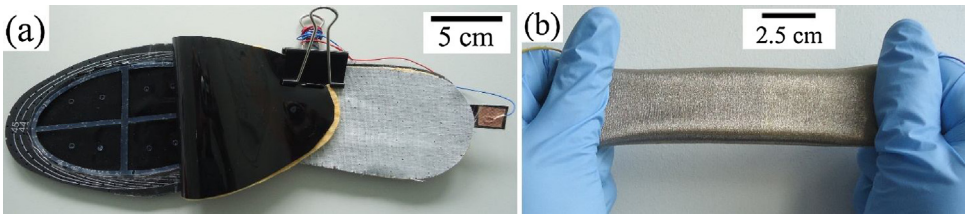

Fig. 4. Photograph of the (a) triboelectric generator based shoe-insole and (b) conductive fabric based stretchable triboelectric band.

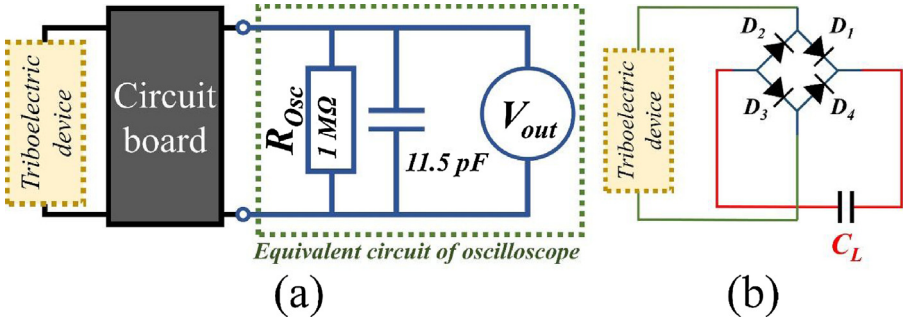

Fig. 5. Schematic of the circuit diagrams for the characterization of (a) electrical responses and (b) amount of charges transferred by triboelectric devices.

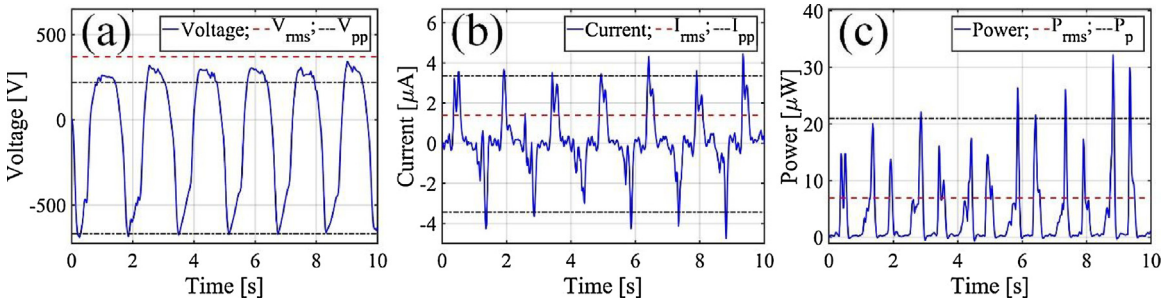

Fig. 6. Electrical responses of the triboelectric generator based shoe insole while walking: (a) open circuit voltage (V_{OC}), (b) maximum output current (I_{Max}) and (c) power generated for the load resistance of 1 M Ω.

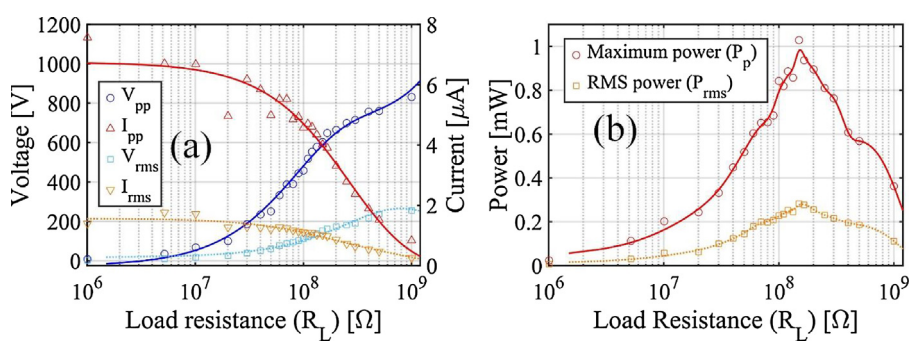

Fig. 7. Electrical responses with respect to the load resistances (R_L) of the triboelectric generator based shoe insole while walking: (a) generated voltages and currents, and (b) output powers.

motions. Thereafter, electrical responses, the charges transferred and the energy stored of the triboelectric stretchable band were finally tested on body by fixing it on elastic hand band and measuring performances during arm stretching motion at 0.65 ± 0.15 Hz.

4. Results and discussions

4.1. Triboelectric shoe insole

During walking, the peak-to-peak and rms V_{OC} of 885 V and 368 V, and I_{Max} of 7.6 μA and 1.4 μA were observed. Figs. 6(a)–(c) present the responses of the shoe insole during walking. Consequently, the voltage, current output responses and resulting power were measured for different load resistances (R_L), as illustrated in Fig. 7. While walking, optimum peak power (P_p) and rms power (P_{rms}) of 1.03 mW and 0.28 mW, that corresponds to peak and rms power density (P_d) of 7.74 μW/cm^2 and 2.13 μW/cm^2 was obtained for the R_L of 150 M Ω. The amount of charges transferred and energy stored in the capacitor for single walking step were 0.90 ± 0.03 μC/cycle and 0.18 ± 0.01 μJ/cycle, respectively.

Figs. 8(a)–(c) demonstrate the output responses, namely, V_{OC}, I_{Max} and power for R_L of 1 MΩ of the shoe insole while run-

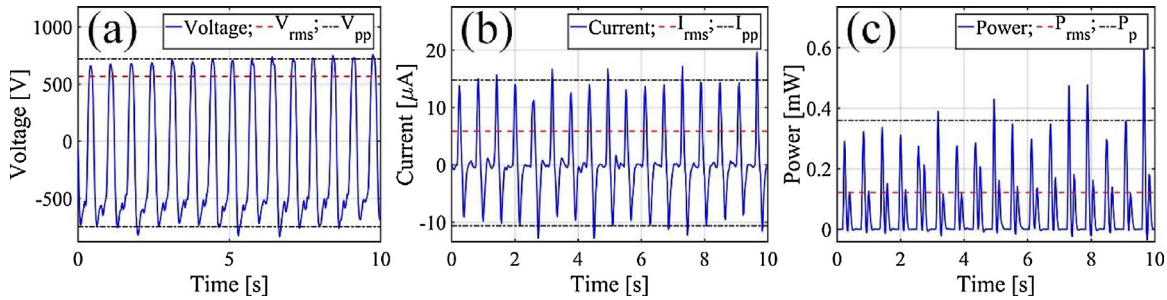

Fig. 8. Electrical responses of the triboelectric generator based shoe insole while running: (a) open circuit voltage (V_{OC}), (b) maximum output current (I_{Max}) and (c) power generated for the load resistance of 1 M Ω.

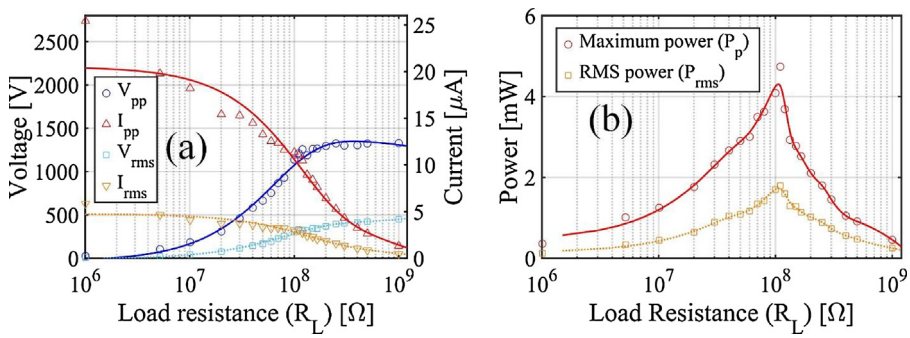

Fig. 9. Responses of the triboelectric generator based shoe insole while running with respect to the load resistances (R_L): (a) voltage and current outputs and (b) output powers.

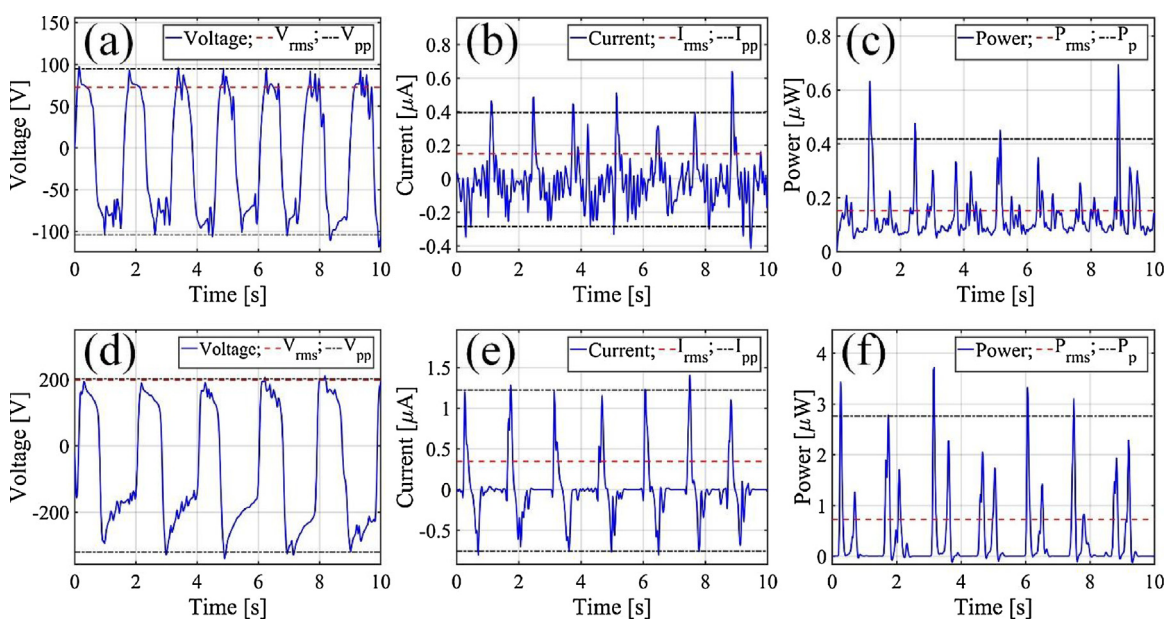

Fig. 10. Electrical responses of the PDMS-PU based stretchable triboelectric band. For stretching mode: (a) V_{OC}, (b) I_{Max} and (c) power output for the R_L of 1 M Ω. For Tapping mode: (d) V_{OC}, (e) I_{Max} and (f) power for the R_L of 1 M Ω.

ning condition. The peak-to-peak and rms value of V_{OC} of 1468 V and 566 V, and I_{Max} of 25.4 μ A and 5.9 μ A were recorded, respectively. In addition, for different resistance values, the voltage and current responses, and corresponding powers, were also studied. Fig. 9 presents the electrical responses with respect to load resistances for running. In this case, the optimum P_p of 4.74 m W and P_{rms} of 1.79 m W was generated by the shoe insole for the R_L of 110 M Ω, which resulting to the peak and rms power density (P_d) of 35.66 μ W/cm^2 and 13.51 μ W/cm^2, respectively. The running effort provided the amount of charges transferred of 1.14 ± 0.06 μ C/cycle and energy stored in the capacitor of 0.30 ± 0.03 μ J/cycle.

As observed, the vertical-contact separation mechanism based shoe insole showed capabilities of converting mechanical energy to electric signal for walking and running movements. The peak-to-peak (*PP*) and root mean square (*RMS*) value of the responses of the shoe insole while different movements are listed in Table 2. As expected, the shoe insole performed better during running, which was associated to the relative higher operational frequency, velocity of kinetics and higher pressure generated while running. Such

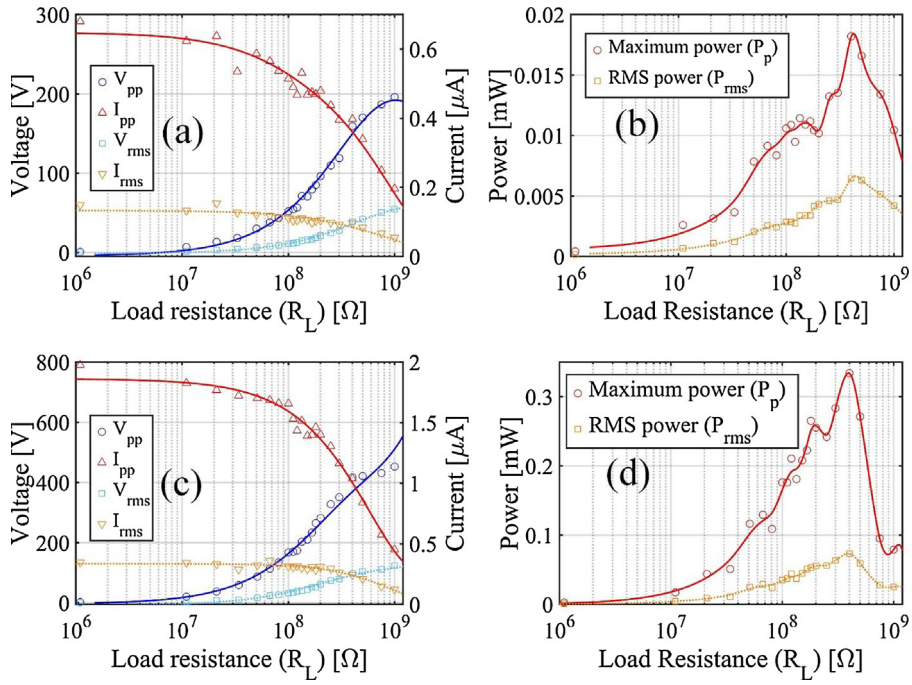

Fig. 11. Electrical responses of the PDMS-PU based stretchable triboelectric band with respect to the load resistances. For stretching mode: (a) voltages and currents generated and (b) corresponding output powers. For tapping mode: (c) voltage and current outputs and (d) corresponding powers.

Table 2
List of responses of the triboelectric shoe insole for walking and running movements.

Responses	Walking		Running	
	PP	*RMS*	*PP*	*RMS*
V_{OC} [V]	885	368	1468	566
I_{Max} [μA] (at 1 MΩ)	7.6	1.4	25.4	5.9
Optimum load resistance [MΩ]	150		110	
P (optimum) [mW]	1.03	0.28	4.74	1.79
P_d (optimum) [μW/cm^2]	7.74	2.13	35.66	13.51
Charge transfer [μC/cycle]	0.90 ± 0.03		1.14 ± 0.06	
Energy stored [μJ/cycle]	0.18 ± 0.01		0.30 ± 0.03	

Table 3
Responses of the stretchable triboelectric band under stretching and tapping conditions.

Responses	Stretching		Tapping	
	PP	*RMS*	*PP*	*RMS*
V_{OC} [V]	203	73	522	198
I_{Max} [μA] (at 1 MΩ)	0.68	0.15	1.98	0.35
Optimum R_L [MΩ]	400			
P (optimum) [μW]	18.2	6.5	333.9	72.6

triboelectric device can be implements on body for energy scavenging, and as well as for motion sensing.

4.2. Stretchable triboelectric band

The developed stretchable band introduced both sliding and vertical-contact separation mechanism within the design. The electrical responses of the conductive fabric based triboelectric band was studied under stretching and tapping conditions to determine the respective optimum load resistances for the operating frequency of 0.65 ± 0.15 Hz, as presented in Fig. 10. The stretching of the triboelectric band produced the rms V_{OC} of 73 V and I_{Max} of 0.15 μA. Whereas, for the tapping mode of the same triboelectric device, the rms V_{OC} and I_{Max} of 198 V and 0.35 μA, respectively, were generated. The responses of the stretchable band with respect to the varying load resistance (R_L) for stretching (Fig. 11(a and b)) and tapping (Fig. 11(c and d)) operations were examined. The optimum rms power outputs of 6.5 μW and 72.6 μW were obtained for stretching and tapping, respectively. The optimum load resistance (R_L) for both characterization modes was 400 MΩ. Table 3 summarized the responses for the stretchable triboelectric band.

Afterwards, the stretchable triboelectric band was stitched on stretchable band (Fig. 12), and sensing and energy harvesting performances of the device was examined, while the arm stretching movement, for the optimum load resistance of 400 MΩ. The

Fig. 12. Photograph of the conductive fabric based triboelectric band mounted on body using stretchable band.

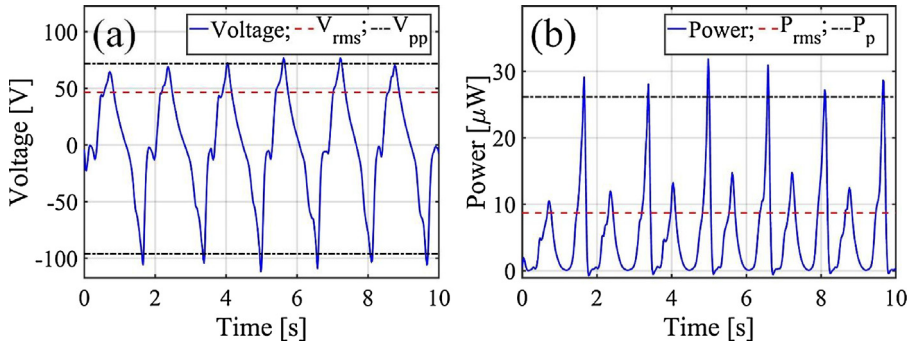

Fig. 13. Responses of the triboelectric band for arm stretching movement: (a) output voltage and (b) power for the R_L of 400 M Ω.

Table 4
Responses of the triboelectric stretchable PDMS-PU band during arm stretching motion.

Responses	Arm stretching	
	PP	*RMS*
Load resistance [M Ω]	400	
Voltage [V]	168	46
Current [μ A]	0.42	0.12
Power [μ W]	26	9
Power density [μ W/cm^2]	0.36	0.12
Charge transfer [n C/cycle]	251 ± 11	
Energy stored [n J/cycle]	14.4 ± 1.2	

stretchable triboelectric band generated the peak-to-peak output voltage (V_{pp}) and current (I_{pp}) of 168 V and 0.42 μ A, and rms voltage (V_{rms}) and current (I_{rms}) of 46 V and 0.12 μ A over 400 M Ω of R_L, that corresponded to the peak and rms power output of 26 μ W and 9 μ W, respectively. Fig. 13 presents the results for the arm stretching motion for the triboelectric stretch band.

For arm stretching, the triboelectric stretchable band provided the amount of charges transferred of 251 ± 11 n C/cycle and energy stored of 14.4 ± 1.2 n J/cycle. The sensing and energy scavenging performances of the triboelectric band are tabulated in Table 4. It has been observed that the use of PDMS and PU elastomers exhibited higher sticking between layer and thus causes higher friction, and thus occasionally brought down the performance of the device. This stickiness between two layers can be reduced by adapting etching process, such as, laser engraving, of the PDMS and PU surfaces.

5. Conclusion

We presented, herein, the pattering of soft, flexible, biocompatible elastomers, like, PDMS and PU, and their carbon based conductive composites using simple casting technique to develop triboelectric devices. The employment of film casting method and elastomeric materials enable fast and cost-effective method to prepare multilayer structures that can easily be adapted for large area applications. The methodology also enabled prompt integration of the elastomer layer with the textile. The next phase of the work is to use commercial PDMS and PU roll to start the process. In that way, we can eliminate the use of PET, sacrificial layer and the deposition of protective layers, which will reduce the fabrication steps. The development approach was tested by preparing vertical-control separation mode triboelectric shoe insole and stretchable triboelectric band. The result showed that the technology exhibited the potential to develop soft triboelectric devices that can be used for sensing of and energy harvesting from low frequency movements. However, the triboelectric devices presented here were the first initiatives toward development of fully casted soft triboelectric energy harvesters and motion sensors. Further advanced studies regarding materials, designs optimization along with mounting on different body location to enhance the feedbacks of the devices and for generating higher amount of charges transferred will be required.

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